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Description

The invention relates to a dimensionally stable laminate in the form of a flat panel, a tube or an unflat three-dimensional shape, and comprises a number of superposed, pressed-together thermoplastic plastic films, that are coated at least on one side with a sealing layer, the melting point of which is lower than the melting point of each of the individual plastics films and which are monoaxially and/or biaxially stretched and a method of manufacture.

Recently, an increased trend has been observed in the use of high-capacity composites in various sectors, such as vehicle construction, aircraft construction, and instrument design. These include, inter alia, thermosetting and thermoplastic polymer materials, whose mechanical properties are considerably improved, in comparison with unreinforced polymers, by the introduction of reinforcement materials in the form of textile fibers, glass fibers, carbon fibers or aramid staple fibers. As an alternative to the introduction of reinforced materials, techniques have been developed to manufacture high-strength laminations by means of the orientation process.

As a technique within the context of the orientation process, the method described in EP A1 – 02 07 047 may apply, wherein a number of biaxially stretched films, fitted with thin, co-extruded sealing layers are pressed together under pressure and temperature into a thick, homogenous laminate.

One method-specific procedure of this technique, i.e., the application of individual plastic films, facilitates the targeted insertion of functional layers at any point of the shaped body and thus the customizing properties of the manufactured shaped body. Thus, for example, biaxially stretched films can be combined with monoaxial or unstretched similar or dissimilar films, in order to influence the fracture behavior of the shaped body in desired ways. However, the manufacturing properties obtained in this manner are insufficient in many cases to handle the extreme mechanical requirements of heavily loaded materials in practice.

In patent AT-PS 3 83 542, a rigid shaped body, such as a panel or a tube, is described as consisting of a large number of superposed and/or adjacently layered, and pressed-together, oriented, i.e., stretched thermoplastic carriers in the form of films, ribbons, strands or fibers. The stretched plastic carriers on the sides touching the structure during the vertical and horizontal stacking of the superposed and pressed-together layers are coated with thermoplastic material, whereby the coating material has a lower crystallite melting temperature than the plastic carrier and the thickness of every two superposed coating materials is lower than the thickness of one plastic carrier.

The method suggested in the Austrian Patent Application AT-PS 3 83 542 produces high-strength, mechanically stable panels by applying pressure on stretched, co-extruded thin plastic films, under the influence of heat, whereby on the opposite contact surfaces, the thermoplastic coated plastic carrier is pressed at a temperature above the crystallite melting temperature of the coating material and below the crystallite melting temperature of the plastic carrier. Based on its underlying principle, this method offers, by pressing at an increased temperature, the possibility of preventing a considerable drop of the targeted advantageous mechanical and physical properties due to the stretching of an individual plastic carrier, in contrast to alternative panel manufacturing technologies, such as extrusion, casting and calendering, so that the shaped body, despite its great dimensions, which far exceed the dimensions of an individual plastic carrier, has approximately the mechanical and technological properties of a single stretched plastic carrier. Since a film staple consisting of individual similar or different films is coated and pressed by definition, targeted functional layers may, on the one hand, be applied either on the surface of the panel or embedded in the form of intermediate layers in the panel, and on the other hand, by selection of suitable pressing matrices, suitable surface structures are formed simultaneously.

The aim of this invention is to further develop a high-strength, dimensionally stable shaped body of the type described at the beginning as a flat, spherically or cylindrically formed or tubular shaped body and to increase its mechanical strength in such a way that the shaped body can be used as a structural semi-finished product.

This task is solved according to the invention, whereby at least one reinforcing layer fitted with open cross-sections is embedded in the composite body, which is enclosed on all sides by a sealable polymer and that the polymer fills the open cross-sections of the reinforcing layer from both sides and forms an inseparable bond with the reinforcing layer.

In a further development of the invention, each of the sealing layers made from the sealable polymer abut the reinforcing layer on each side, and the open cross-sections of the reinforcing layer are pores, and the polymer of the sealing layers fills the pores of the reinforcing layer from both sides, forming an inseparable bond with the reinforcing layer.

According to the design of the invention, the individual reinforcing layer is incorporated in the middle of the composite body and encloses each of the two partially touching sealing layers that are attached to the adjacent plastic films, with the reinforcing layer along its surface structure.

Another design of the invention results in the patent claims 4 to 17.

The method for manufacturing a composite body by pressing a film staple from superposed, biaxially and/or monoaxially stretched, thermoplastic, co-extruded plastic films is designed such that, before pressing in the film staple at any point, at least one reinforcing layer in the form of fibrous filaments of a non-woven fabric, or of a woven fabric, a crocheted fabric, a knitted fabric or of fleece is embedded in such a way that it lies within the boundaries of the cutting of the plastic films, and that, subsequently, a thermal pressing is carried out between two pressing matrices, which may have either smooth and/or textured surfaces, and that the sealing layers transferred in the molten state to the plastic films both enclose the reinforcing layer on all sides as well as fill its open cross-sections or pores, so that once the film staple has cooled off, an inseparable bond is formed by the plastic films, sealing layers and the reinforcing layer.

Films made of polypropylene and polyester are preferably used for the manufacturing of the composite body, although other suitable polymer materials or polymer combinations may well be used. Polypropylene and polyester films are noted for their low manufacturing costs, and, in addition, they exhibit a high mechanical strength when they are biaxially or monoaxially stretched. Such a composite body, manufactured by the pressing of such a film staple, which comprises a number of biaxially stretched plastic films layered in the same direction or a different direction, and fitted with outer sealing layers, as well as at least one reinforcing layer incorporated within, which consists of textile fibers, carbon fibers, aramid stable fibers or fibers made of liquid crystalline polymers or surface structures in the form of non-woven fabrics, woven fabrics, crocheted fabrics, knitted fabrics or fleece form a structural semi-finished product of good value and with high tensile strength.

The invention is explained in more detail below by means of illustrated embodiments. Shown are:

Fig. 1 A schematic cross-sectional view of a first embodiment of a flat, lamellar composite body according to the invention

Fig. 2 A schematic cross-sectional view of a second embodiment of a flat lamellar composite body, with a thicker reinforcing layer than the first embodiment according to **Fig.** 1.

Fig. 3 A schematic cross-sectional view of a third embodiment of a flat, lamellar composite body, with a reinforcing layer that is completely embedded in a sealing layer film.

Fig. 4 A schematic cross-sectional view of a fourth embodiment of a curved, lamellar composite body with several reinforcing layers.

Fig. 5 A schematic cross-sectional view of a fifth embodiment of a flat, lamellar composite body with several reinforcing layers, which only stretch over partial areas within the composite body.

Fig. 6 A top view of a cutting of a single reinforcing layer, which consists of a fibrous fabric and

Fig. 7 A cross- section of a tubular composite body according to the invention.

A first embodiment of a flat, lamellar composite body 1 is illustrated in **Fig.** 1 and consists of a polymer matrix 2, in the middle of which a reinforcing layer 3 is embedded. The polymer matrix 2 consists of a sealable polymer enclosing the reinforcing layer 3 on all sides, which consists of a fibrous fabric, for example, of which, for the purposes of better clarity in the section, only the warp and weft threads are shown. The polymer matrix 2 is manufactured by a plastic film staple 4, 4' pressed at a pre-determined temperature. As can be seen from the enlarged cutout in **Fig.** 1, the thickness of the fibrous fabric and the enveloping sealing layer 5, 5' is approximately in the order of magnitude of the thickness of a single plastic film 4 or 4'. On each plastic film 4, 4', there is at least one sealing layer 5 and 5' made of a sealable polymer on one side. The thickness of the sealing layers 5 and 5', which are generally hot sealing layers that are attached to the plastic films or carrier films 4, 4', sufficiently embed the reinforcing layer 3 on all sides and form a sufficiently good bond in the boundary layers between the sealing layers and the fibrous fabric of the reinforcing layer. During the pressing method with heat application on the plastic film staples, in which the reinforcing layer 3 made of fibers or fibrous semi-finished products is embedded, the polymer of the sealing layers 5, 5' turns into a molten state, so that the open cross-sections or pores of the reinforcing layer 3 are filled from both sides and, as a result, penetrate through them, in order to form an inseparable bond between the reinforcing layer 3 and the polymer matrix 2.

The sealing layers 5, 5' generally have thicknesses in the order of 1 μm. In **Fig.** 1, the plastic films with their sealing layers, which lie above the reinforcing layer 3, are identified by reference numbers without lines and the corresponding plastic films with their accompanying sealing layers, which are located beneath the reinforcing layer 3, are identified by reference numbers with lines.

Each of the two partially touching sealing layers 5, 5' that indirectly border the reinforcing layer 3 in **Fig.** 1, and which are attached to adjacent plastic films, enclose the reinforcing layer along its surface structure.

Fig. 2 shows a second embodiment of a composite body 1, which is constructed similar to the composite body in **Fig.** 1. The top surface, including the top-sealing layer 5 of the composite body 1 is textured, as shown in the drawing, while the bottom surface of the composite body 1 is smooth. Obviously, both the top surface as well as the bottom surface may be either smooth or textured. The reinforcing layer 3 and the fibrous fabric used for it are thicker than the reinforcing layer according to **Fig.** 1. Consequently, two thicker hot sealing layers 5, 5' are inevitably required on both the plastic films or carrier films 4, 4' forming the respective boundary layers of the reinforcing layer 3. Both the plastic films 4, 4', which form boundary layers of the reinforcing layer 3 differ from the other plastic films 4, 4' of the composite body 1 or of the plastic film staples only in that their sealing layers lying in direct contact with the fibrous fabric of the reinforcing layer 3 have a considerably greater layer thickness than their sealing layers 5, 5', which are turned away from the reinforcing layer 3.

With a third embodiment of the composite body 1, as shown in **Fig.** 3, a very thick reinforcing layer 3 varying in the mm range is incorporated. To this, a thick hot sealing layer in the form of a self-supporting sealing layer film 6, which consists of the same polymer material as the sealing layers 5, 5' of the plastic films 4, 4', is introduced into the boundary layer formed by the fibrous fabric of the reinforcing layer 3 of the

composite body 1. The thickness of the sealing layer 6 is approximately twice the thickness of the reinforcing layer 3. The mass of the sealing layer film 6 must be sufficient to fill the open cross-sections or pores of the fibrous fabric of the reinforcing layer 3 during the pressing process and to form a bond with the adjacent sealing layers 5, 5'. The top and bottom surfaces of the composite body 1 each form textured plastic films 4 and 4'. When necessary, the reinforcing layer 3 can also be embedded between several sealing layer films, coated on both sides, if the thickness of the reinforcing layer 3 requires it. The sealing layer films 6 are generally unstretched, amorphous, tough-elastic polymer layers, which, due to their tough-elastic properties, considerably increase the tensile strength when embedded in the composite body 1.

The self-supporting sealing layer film 6 consists of the same polymer material as the sealing layers 5, 5' of the plastic film or carrier films 4,4'; however, it is also possible for the sealing layer film 6 to be formed from another polymer material than that of the sealing layers 5, 5'.

The sealing layer film 6 may be inserted either as a single film with superposed layers of the film staple, or as a hot sealing layer 6, which has already been reinforced by fibers introduced into the staple. The reinforcing layer 3 then forms a fiber layer, which is embedded in the self-supporting sealing layer or the adhesion layer film 6. The bonding of the fiber layer, sealing layer or adhesion layer then forms a separate layer component, which is embedded in the film staple of the composite body 1. The fiber-reinforced sealing layer film 6 is achieved in such a way, for example, that the fiber reinforcing material is inserted between two hot sealing layer sheetings, and this three-layer bond is moved through the double layer press, in which the fusing sealing layers of the fibrous fabric soak under pressure and temperature and form a bond from pre-impregnated material.

In **Fig.** 4, a schematic sectional view of a fourth embodiment of a curved, lamellar composite body 1 is shown, which contains several reinforcing layers 3, 3', 3". The reinforcing layers 3, 3', 3" have either the same distance or different distances between them, whereby, the orientations of the materials of the reinforcing layers may also be the same or different. The top surface and the bottom surface of the composite body may be both smooth or both of them may be textured, and likewise, it is possible that one of the surfaces is smooth and the other is textured. In another form of this embodiment, at least one of the reinforcing layers may form one of the outer surfaces of the composite body. The composite body is not limited to a curved panel in its shaping, but it may take any unflat three-dimensional shape.

Fig. 5 shows a slightly modified fifth embodiment across from figure 4 of a flat lamellar composite body 1 with several reinforcing layers. Two reinforcing layers 3 and 3' are each stretched over two partial areas of the composite body 1, while another reinforcing layer consists of several partial area layers 3a, which are embedded spaced apart in the same plane of the composite body 1. The two partial areas of the reinforcing layers 3, 3' may be arranged, for example, symmetrically in the middle of the composite body 1. The partial area layers 3a are arranged in the central plane of the composite body, but they may also be embedded in a plane that is at a distance from the composite body.

A top view of an individual reinforcing layer 3, which consists of a fibrous fabric, is shown in **Fig.** 6. The open cross-sections or pores 7 of the fibrous fabric can be clearly recognized in this top view of a section.

Fig. 7 illustrates a cross-section of a tubular composite body 8 according to the invention. This tubular composite body is obtained by a longitudinal winding on a mandrel of biaxially stretched, thermoplastic, co-extruded plastic films 4, 4', which are fitted with sealing layers 5 and 5', whereby the winding of the film layers between these fibrous filaments, non-woven fabrics, woven fabrics, crocheted fabrics, knitted fabrics or fleece are inserted as part of the reinforcing layers. These types of fiber-reinforced tubular composite bodies 8 are bending-resistant and high-strength structural

elements in scaffolding and superstructure construction. Tubular composite bodies 8 reinforced with electroconductive fibrous layers can be used as heated transport systems to extract cold-sensitive liquids, such as oil.

As has already been mentioned, the plastic films of the composite bodies 1 and 8 concern thermoplastic films from the polyester and polyolefin group, whereby each individual plastic film is fitted with at least one hot sealing layer 5 and 5'. Polyester, homopolymers and copolymers, mixtures of various polyesters as well as blends of polyesters with other polymers belong to the group of polyesters. This group includes, in particular, polyethylene terephthalate, polyolefin, as well as polypropylene, polyethylene, polystyrol, polyamide and polyvinyl acetate. In addition to copolymers, terpolymers or blends of these are envisaged as material for the plastic films. As polyolefins, particularly propylene-homopolymers or copolymers are used, whereby the majority of the latter are composed of propylene units.

The plastic films 4, 4' generally have a thickness in the range of 10 to 300 μm, in particular, from 40 to 200 μm.

The thicknesses of the hot sealing layers 5, 5' with one or both sides attached to the plastic film or carrier film, customarily do not exceed 10 % of the thickness of the plastic film, and generally lie in the range of 0.5 μm to 5 μm. These sealing layers 5, 5' consist of inter alia, statistical copolymers or terpolymers of α-olefins with 2.3 and/or 4 C-atoms, copolyesters with ethylene terephthalate units and isophthalate units, ethylene vinyl acetate copolymers, acrylate copolymers, copolyamides, polyurethanes, and polyvinyl butyral.

Textile fibers, glass fibers, carbon fibers and aramid staple fibers or fibers consisting of liquid crystalline polymers, as well as surface structures consisting of the above-named fibers in the form of non-woven fabrics, woven fabrics, crocheted fabrics, knitted fabrics

or fleece are used for the reinforcing layers of the composite body. Reinforcing layers made of combinations of these fibers or surface structures made of these fibers are also taken into consideration. A composite body may be made of the same materials or different materials.

The reinforcing layers of The fibers or surface structures made of fibers can be prepared on the surface for better bonding in the polymer matrix 2 of the composite body and for increasing the interlaminar shearing strength. In addition to the shearing strength, this surface preparation improves the adhesive strength of a reinforcing layer in the polymer matrix. For example, a polyamide impregnation or waterproofing of a reinforcing layer is feasible if the reinforcing layer is run through a polyamide solution before its incorporation in the polymer matrix. If it is desired that the composite body have electrical conductivity, metallized fibers or fibrous semi-finished products are used as reinforcing layers.

Although it is not illustrated, instead of bonded fibers or surface structures made of corresponding fibers, it is also possible to embed single filaments in the polymer matrix in area-measured order and with corresponding density.

The manufacturing of the lamellar composite body is achieved by pressing a film staple from superposed, biaxially and/or monoaxially stretched, thermoplastic, co-extruded plastic films. Prior to the pressing process, at any point in the plastic staple, at least one reinforcing layer in the form of fibrous filaments, of a non-woven fabric, of a woven fabric, of a crocheted fabric, of a knitted fabric, or of fleece is incorporated in such a way that it lies within the boundaries of the cutting of the plastic films. If only one reinforcing layer is envisaged, then it is generally located in the middle in the film staple. If several reinforcing layers are introduced into the film staple, then at least one of these reinforcing layers may be located on an outer surface of the film staple. The spacing between the individual reinforcing layers may be the same or this spacing may vary. The thermal pressing of the plastic staple takes place between two pressing matrices, which

may have smooth and/or textured surfaces. The sealing layers in the molten state transferred to the plastic films enclose the reinforcing layer(s) on all sides and fill its open cross-sections or pores in such a way that, once the film staple has cooled off, an inseparable composite body made of plastic films, sealing layers and reinforcing layer(s) is present in the polymer matrix.

The composite bodies reinforced by the embedding of fibrous layers are know for their outstanding tensile strength and are used, in particular, for the manufacturing of heavily loaded mechanical components.